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Radiation effects on InGaN quantum wells and GaN simultaneously probed by ion beam-induced luminescence

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Abstract

InGaN quantum well structures on GaN epilayers were exposed to 500 keV alpha particles to fluences above 10^{14} cm^2 to probe the relative radiation tolerance of the epilayer and wells. Performance was quantitatively estimated by the intensity of ion-beam induced luminescence. Two separate types of quantum well structures emitted at 470 and 510 nm prior to irradiation, and only small wavelength shifts were observed even with the highest alpha fluences. Complementary cathodoluminescence experiments quantitatively showed that luminescence in the quantum wells is strongly influenced by charges injected deep into the GaN epilayer. The 500 keV alpha penetration depth was ~ 1 μm, ensuring a higher defect production rate in GaN than InGaN as alpha particles slowed and stopped within a minority carrier diffusion length of the quantum wells. Nevertheless, the rate of luminescent decay was similar for both materials. Taken together with the cathodoluminescence data, this indicates that the quantum well luminescence decay rate is dominated by radiation-induced defects in the GaN epilayer. InGaN quantum wells are demonstrated to be not more than a factor of ten more radiation sensitive compared to GaN, and may be substantially better.

Introduction

InGaN quantum well structures can be used as the active region of efficient light-emitting diodes, especially in the wavelength range spanning 400-520 nm, where applications including optical communication systems and components in white light sources are being considered.[1] However, to allow operation of these devices in space and other radiation-rich environments, it is important to understand and control the effects of radiation-induced defects. Previous studies have employed penetrating particles such as neutrons and high-energy protons to probe the radiation tolerance of InGaN light-emitting diodes (LED's)[1] and GaN double-heterojunction light-emitting diodes (LED's)[2], but these experiments have necessarily modified not only the quantum well structures but also the thick GaN epilayer immediately underneath. The measured performance limitations therefore reflect the combined radiation tolerance of the composite system without providing a direct measurement of the light-generating InGaN quantum well layers.

In this work, we perform *in-situ* measurement of ion-beam induced luminescence to simultaneously observe the properties of a five-layer stack of InGaN quantum wells and its GaN epilayer when these materials are exposed to 500 keV alpha particles. We spectroscopically separate emissions from quantum wells and GaN, enabling a more independent estimation of radiation tolerance of the two structures. Next, we perform complementary electron-beam experiments and modeling to quantitatively determine the recombination properties of excited electrons as a function of particle penetration depth. Finally, we model our *in-situ* luminescence results using Monte Carlo simulations of alpha induced lattice defect creation coupled with finite element drift-diffusion simulations.

Experimental details

Quantum wells on GaN epilayers were grown by metalorganic chemical vapor deposition on sapphire substrates. Our tested structures consisted of five bilayer periods of 10 nm GaN and 3 nm of $In_xGa_{1-x}N$ (x between 0.1 and 0.2), on top of ~4 microns of GaN. Two separate microstructures were examined. In the first, with $In_xGa_{1-x}N$, $x \approx 0.1$, there was a high density of lateral inhomogenities such as V-defects, as determined by transmission electron microscopy and atomic force microscopy[3] [4]. The primary photoluminescence peak was at 470 nm. In the second sample, with $In_xGa_{1-x}N$, $x \approx 0.2$, smooth and homogeneous quantum well structures were concluded from well resolved x-ray diffraction patterns and very low growth surface morphology as identified in atomic force microscopy. For this sample, the primary photoluminescence peak was at 510 nm.

Ion beam-induced luminescence experiments were performed at the Lawrence Livermore National Laboratory (LLNL) 4 MeV Ion Beam Accelerator. Samples were mounted on a water-cooled aluminum plate at 45 degrees to the incoming beam of 500 keV alpha particles, which was incident on the ~ 4 cm^2 samples at a current of 13 nA over a rectangular area of 14 mm by 3 mm. The resulting energy deposition rate, about 15 mW/cm^2, is not sufficient to generate a significant temperature increase in the quantum wells or epilayer during irradiation. Light was captured by a 5 mm diameter collimating lens positioned about 1 cm away from the sample, and transported via optical fiber to a spectrometer. In complementary electron beam experiments, the same light collection and spectral analysis system was employed, but light was generated by a rastered electron beam where current varied from 7 to 37 nA, with electron energies between 1 and 30 keV.

Results

Figure 1(a) shows several ion-beam induced luminescence spectra for the $In_{0.1}Ga_{0.9}N$, defect-rich quantum well structure, obtained sequentially as the sample was irradiated. The spectrum for the more homogeneous $In_{0.2}Ga_{0.8}N$ sample, Fig. 1(b), is qualitatively similar. However, some differences, such as the rate of luminescence decay, are discussed in detail below.

Figure 2 shows the electron beam-induced luminescence spectra from the same sample shown in Fig. 1(a). In contrast to the single alpha particle energy used in the ion-beam experiments, the electron energy was varied from 1 to 30 keV to provide insight into the relative radiative recombination efficiencies as a function of electron depth for the quantum well structure and underlying GaN. These keV electrons do not have sufficient energy to cause significant damage in the sample even after many minutes of continuous irradiation. The inset shows the ratio of the quantum well and GaN peak intensities as a function of electron beam energy. There is a small blue shift in the peaks with increasing electron energy. The spectrum for the more homogeneous $In_{0.2}Ga_{0.8}N$ sample (data not shown) is qualitatively similar.

Discussion

Diffusion length and recombination

Fig. 1 demonstrates that the InGaN quantum well region emission is many times more intense compared to emission from the GaN bulk, independent of fluence and sample microstructure. Because the total thickness of the quantum well layers is just 65 nm, it is unlikely that the quantum well signal is due to a re-absorption process of GaN luminescence. However, the diffusion length of minority carriers in GaN is large: 3.4 to 1.2 microns in the doping range of 5×10^{15} – 2×10^{18} cm^3.[5] Charges created by alpha-particle induced ionization in GaN can therefore readily diffuse to the quantum wells at the surface, where they radiatively recombine much more efficiently than in GaN.

In Fig. 1(a), the emission peak at 370 nm is attributed to band-to-band recombination in GaN, while the maximum at 470 nm is associated with emission from the $In_{0.1}Ga_{0.9}N$/GaN quantum wells. In Fig 2(b), the peak at 500 nm is associated with emission from the $In_{0.2}Ga_{0.8}N$/GaN quantum wells. In both spectral series the broad shoulder peaked at 580 nm ("yellow band") is likely created by defect states in the GaN.[5] This broad defect-related peak appears in the electron beam-induced spectra more prominently with increasing electron energy (and therefore penetration depth), indicating that it is indeed associated with defects in the underlying GaN, and not the quantum wells or with alpha-induced defects. Note also the slight blue shift which occurs for all the peaks with increasing fluence, which is possibly attributable to sample charging.

The relationship between emission and charge injection depth is more quantitatively demonstrated by results from electron-beam induced emission experiments (Fig. 2), and by the electron penetration depth calculations shown in Fig. 3. Here penetration depth is taken as the location where an increasing acceleration voltage results in the highest differential energy deposition At 20 keV, which corresponds to a ~3:2 ratio of quantum well to GaN emission signal, the electron penetration depth is about 710 nm – more than 10 times the total thickness of the quantum well stack.

Radiation tolerance

The depth distribution of generated vacancy defects and deposited ionization energy was simulated using the SRIM-2006 code.[7] Fig. 4(a) shows the vacancy profile for 500 keV alpha particles incident at 45 degrees on to the GaN surface. The deposited ionization energy provides the carrier excitation which subsequently is observed by radiative recombination. We assume a linear scaling of the rate of radiative recombination and ionization rate. Therefore, the simulation can be compared directly to the luminescence profile. Fig. 4(b) gives the energy band diagram of the piezoelectric quantum well structure along with the ionization rate as a function of position.

Figs. 4(a) and 4(b) also show the quantum well structure thickness schematically superimposed over the calculated ionization and vacancy profiles. This emphasizes that most of the ionization energy (Fig. 4(b) blue circles) from the 500 keV alpha particles is deposited in the underlying GaN, and that vacancies, which are associated with optical performance loss (Fig 4(a) blue triangles), are predominantly generated there as well.

As shown in Fig. 4(a), the concentration of Ga and N vacancies were calculated as a function of alpha particle fluence from SRIM.[7] From Goodman et al. [8], these vacancies are the primary electrically-active defects generated when GaN is exposed to energetic alpha particles. The N vacancy is an electron donor with energy 0.2 eV below the conduction band of GaN, while the Ga vacancy is an electron acceptor with energy 0.95 eV below the conduction band.[8] The capture cross section of these defects is not well-established, but values around $3x10^{-15}$ cm^2 have been reported.[9].

Using the spatial distribution of Ga and N vacancies estimated from SRIM and the energies of these defects taken from [8], we calculated the radiative and non-radiative recombination rates of electron-hole pairs with Silvaco Atlas.[10] Non-radiative recombination is modeled with trap-assisted recombination and with Shockley-Read-Hall (SRH) recombination, assuming the vacancies act as traps with the appropriate energies. For any point within the quantum well/GaN structure, time dependent recombination rates were calculated as a function of trap density, as shown in Fig. 5. Finally, by spatial and temporal integration, the total radiative recombination in the quantum well/GaN structure was calculated and directly compared to the spectroscopically-measured values obtained as a function of fluence. The capture cross section of the traps could be varied to fit the experimental data. This comparison is shown in Fig. 6.

In Fig. 6, the measured quantum well and GaN contributions were taken from the peak values of alpha-luminescence spectra, shown in Fig. 1. The measured data for both quantum well samples are consistent with a trap cross section with value ~1×10^{-15} cm2, which agrees relatively well with published values [11]. As expected from SRIM, the model confirms the faster degradation rate of GaN vs. the quantum wells due to the larger number of defects introduced in GaN at the end of range of the alpha particles

In previous studies[1, 2], more penetrating particles were used to create defects in light-emitting diodes. For example 40 MeV protons, with a range of approximately 50 μm in GaN, were used to probe InGaN LEDs [1]. In this case, the rate of vacancy creation as a function of depth was constant in the top electrically active approximately 3-5 μm of the InGaN/GaN. The observed performance degradation rate could be dominated either by the GaN or by the quantum wells, but the data did allow independent determination of the relative radiation tolerance of the two materials.

This work, by contrast, demonstrates that the radiation-induced LED performance decay may have been influenced significantly by damage in the GaN epilayers. In our experiments, the spectroscopically-separated epilayer and quantum well emissions enable us to bound the relative performance of the quantum wells vs. GaN. For example, Fig. 4(a) (right axis) shows the alpha particle-induced damage rates quantitatively: in GaN, vacancies are created at an average rate of ~0.01 vacancies/Å/ion, while in the quantum wells, vacancies are created ten times slower - a rate closer to 0.001 vacancies/Å/ion. However, in Fig. 6, we see that the luminescence decay rates for GaN and InGaN quantum wells are similar. This demonstrates that the quantum wells are not worse by more than a factor of 10 compared to GaN, or else this would be reflected in a larger quantum well luminescence decay rate.

Conclusions

InGaN quantum wells and GaN epilayers were probed with 500 keV alpha particles and 1-30 keV electrons to determine their relative radiation tolerance via luminescence. A Monte Carlo model indicates ten times fewer defects were created in the quantum wells, compared to the GaN. However, radiation-induced luminescence decay rates were similar in both materials. Further, the relative alpha- and electron-induced luminescence in the InGaN and GaN demonstrates that most charges injected in the GaN recombined radiatively in quantum wells. Taken together, these observations mean that the luminescence decay rate in the quantum wells was dominated by defects generated in the GaN epilayer. The radiation sensitivity of InGaN quantum wells under our experimental conditions is therefore not greater than a factor of 10 compared to GaN, and it could be substantially smaller.

Acknowledgement

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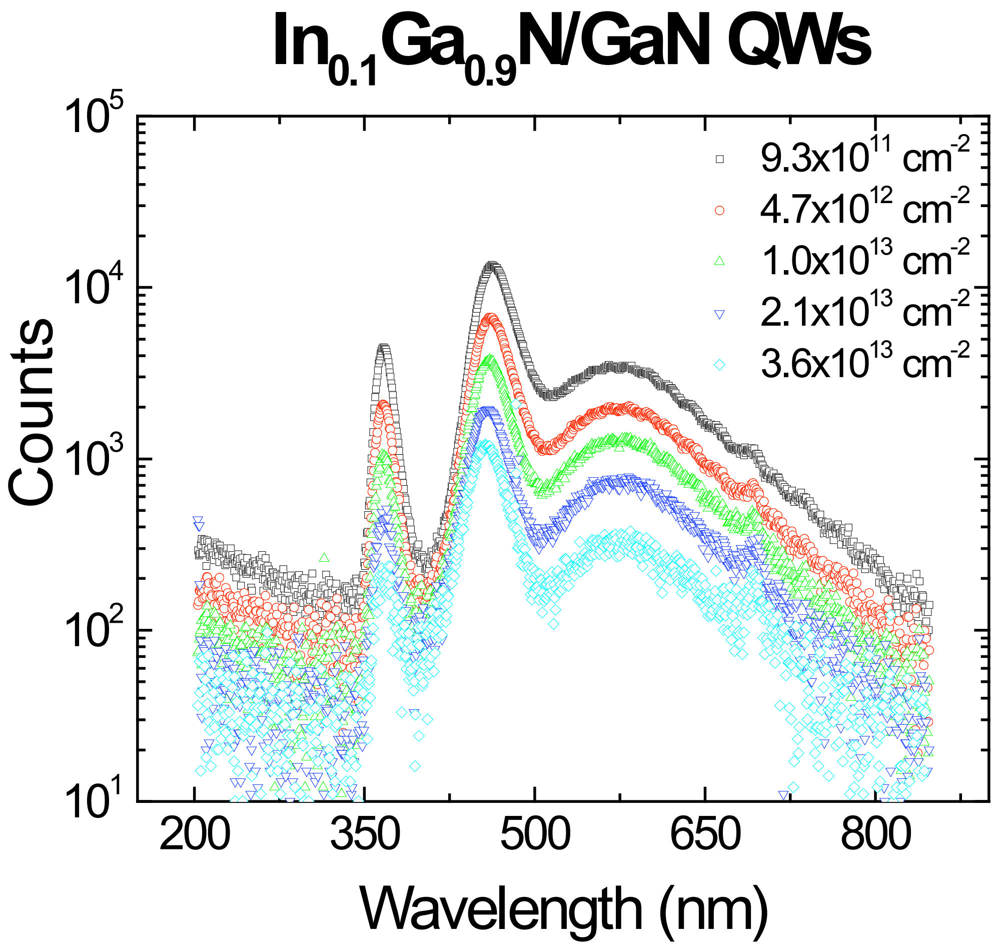
Figures


In0.1Ga0.9N/GaN QWs
10^5
10^4
10^3
10^2
10^1
Counts
200
350
500
650
800
Wavelength (nm)
9.3x10^11 cm^-2
4.7x10^12 cm^-2
1.0x10^13 cm^-2
2.1x10^13 cm^-2
3.6x10^13 cm^-2

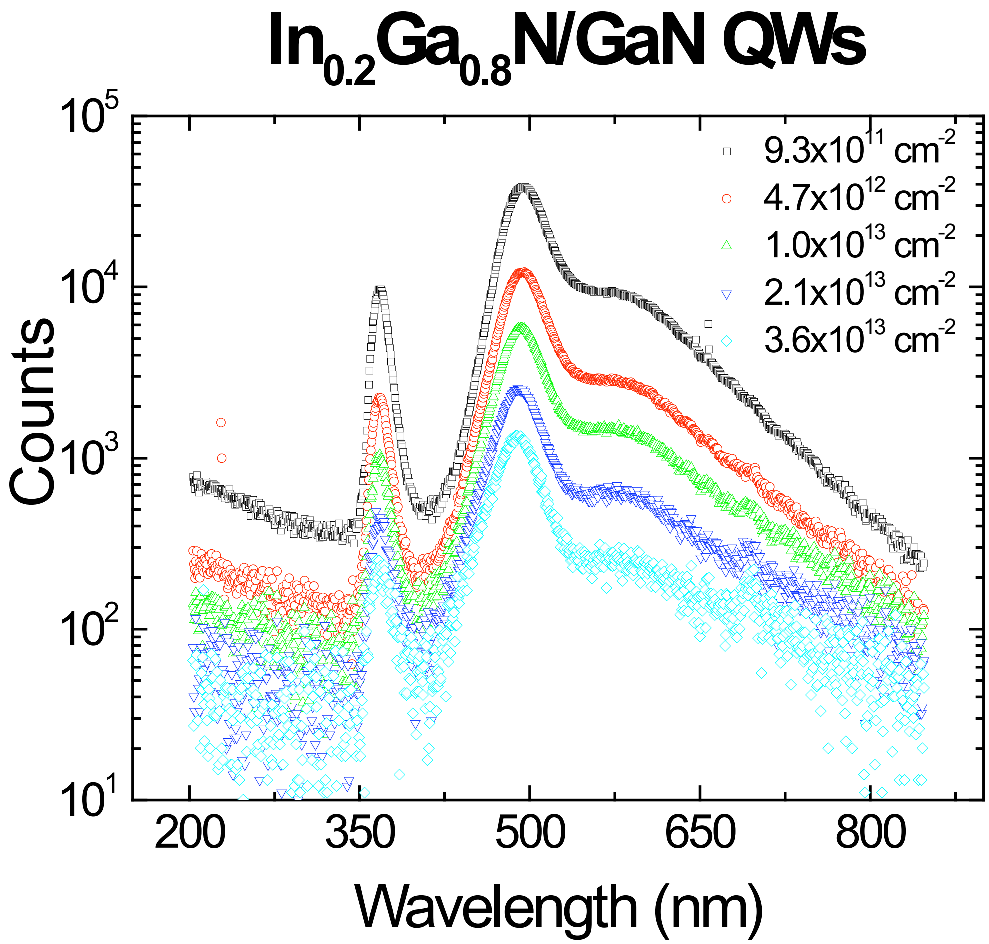


Fig. 1. Ion-beam induced luminescence spectra, obtained sequentially during sample irradiation to a fluence of 1.7×10^{14} alpha/cm^2. Emissions at 470 nm (a) and 510 nm (b) correspond to luminescence from quantum wells; emission at ~365 in both samples is due to luminescence from GaN epilayer.

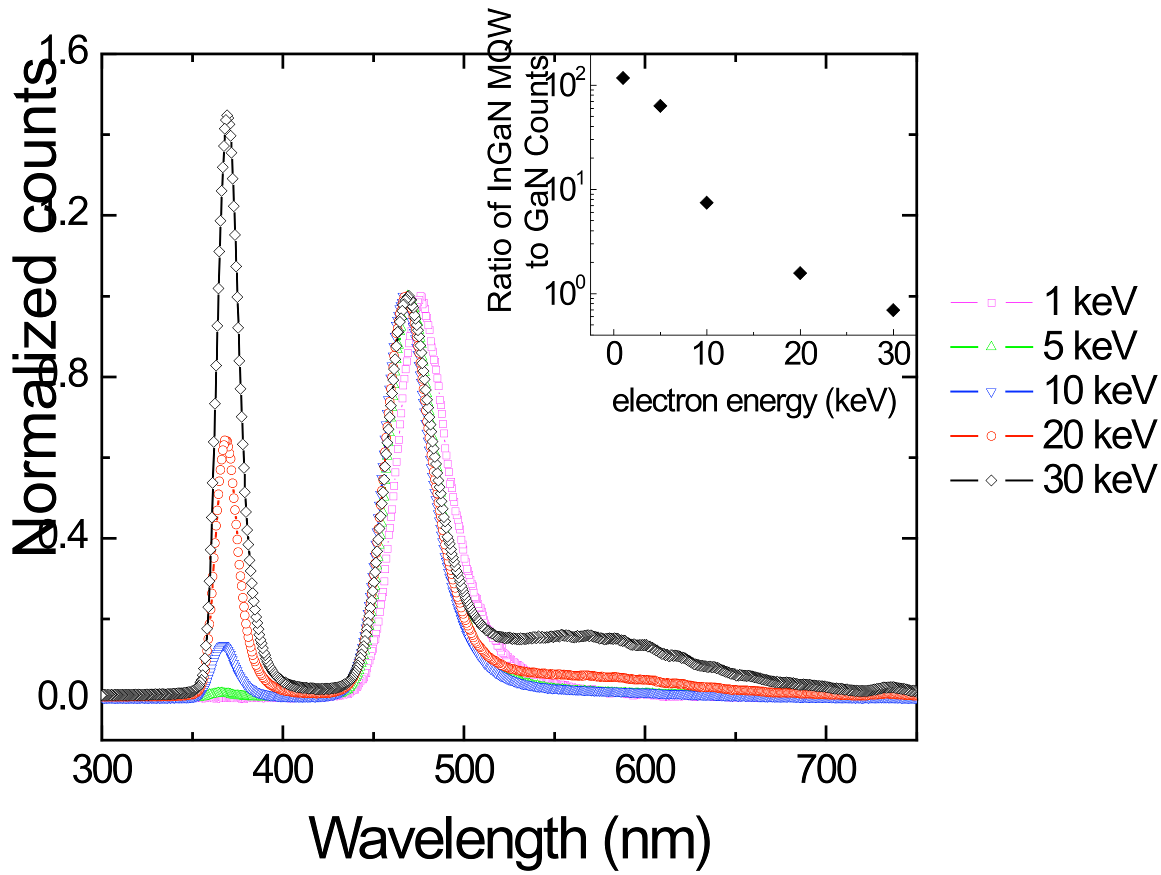


Fig. 2 Spectra, function of electron energy; inset, ratio of InGaN intensity to GaN intensity, function of electron beam energy.

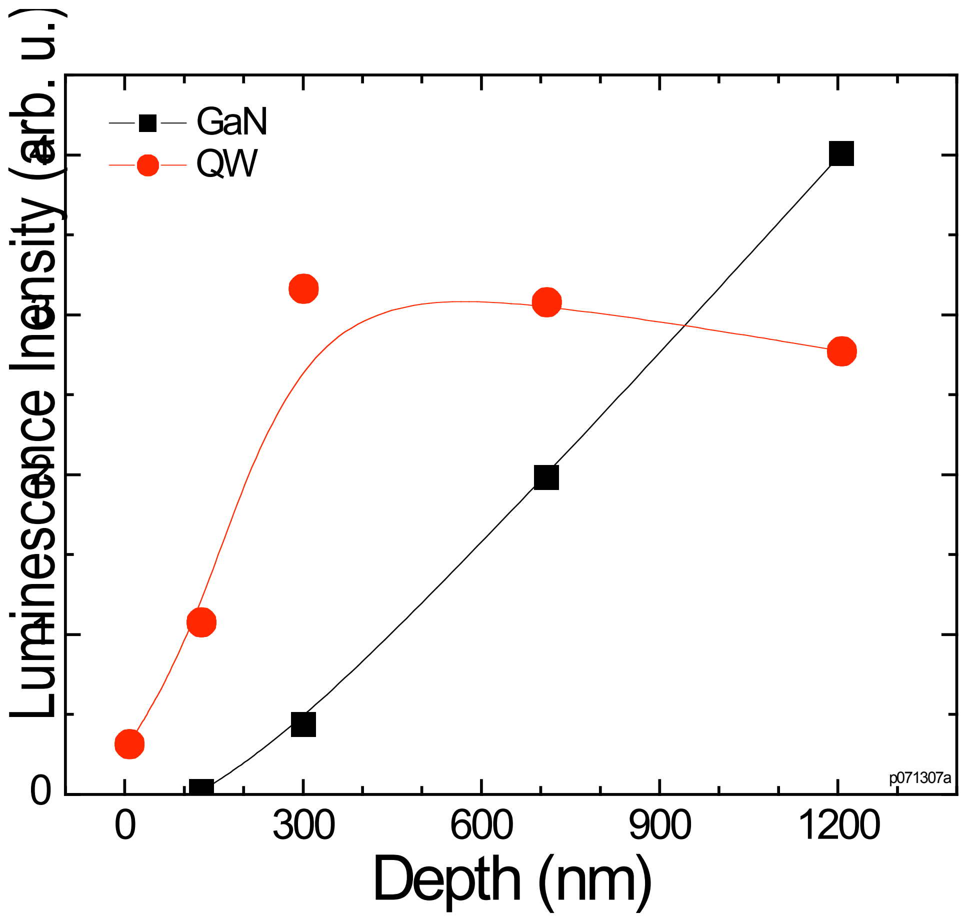
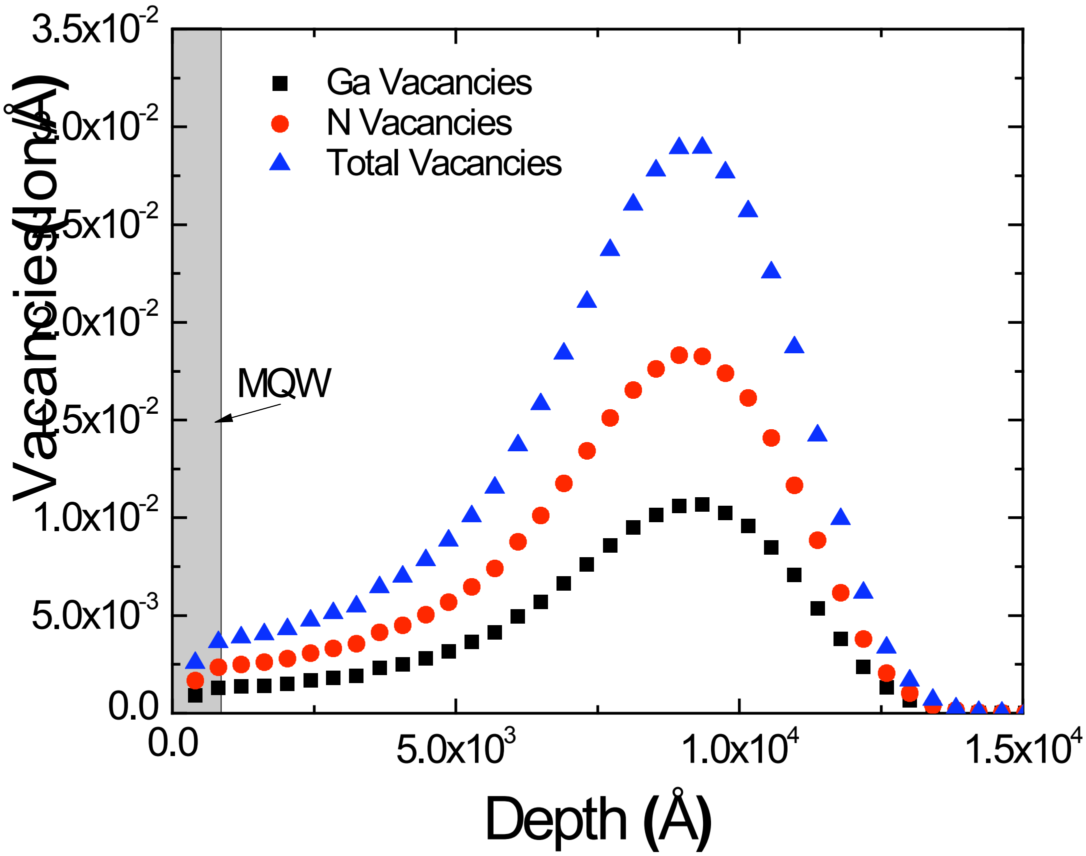
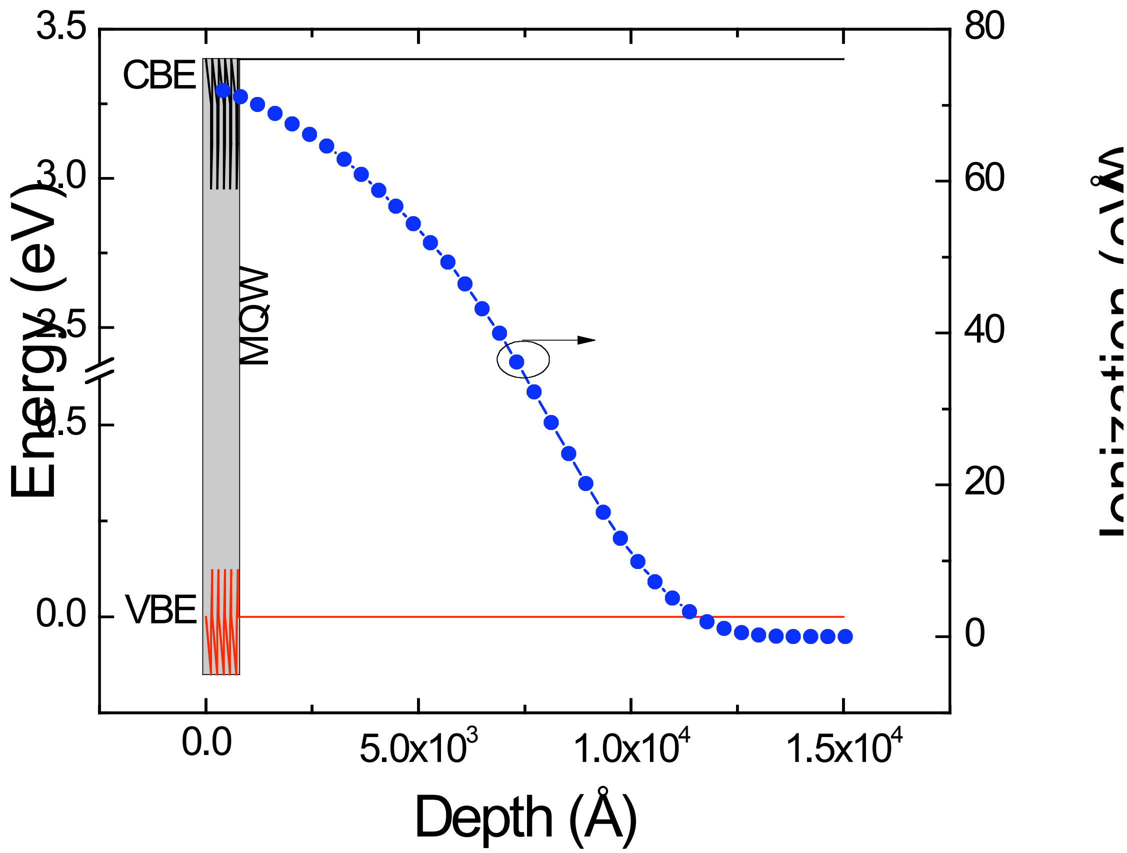


Fig. 3 Calculated CL intensity as a function of penetration depth of the electron beam for GaN/ $In_{0.1}Ga_{0.9}N$ quantum wells. The QW intensity rises rapidly and saturates, while the GaN signal grows steadily (drawn lines are guides to the eye).



Ga Vacancies
N Vacancies
Total Vacancies
MQW
Vacancies (Ion/Å)
Depth (Å)
0.0
5.0x10^3
1.0x10^4
1.5x10^4
5.0x10^-3
1.0x10^-2
1.5x10^-2
2.0x10^-2
2.5x10^-2
3.0x10^-2
3.5x10^-2

a)



b)

Fig. 4(a) SRIM calculation for vacancy creation as a function of depth by alpha particles with 500 keV energy and 45^{o} incidence angle; (b) Energy band diagram of QWs on GaN and ionization rate as a function of depth..

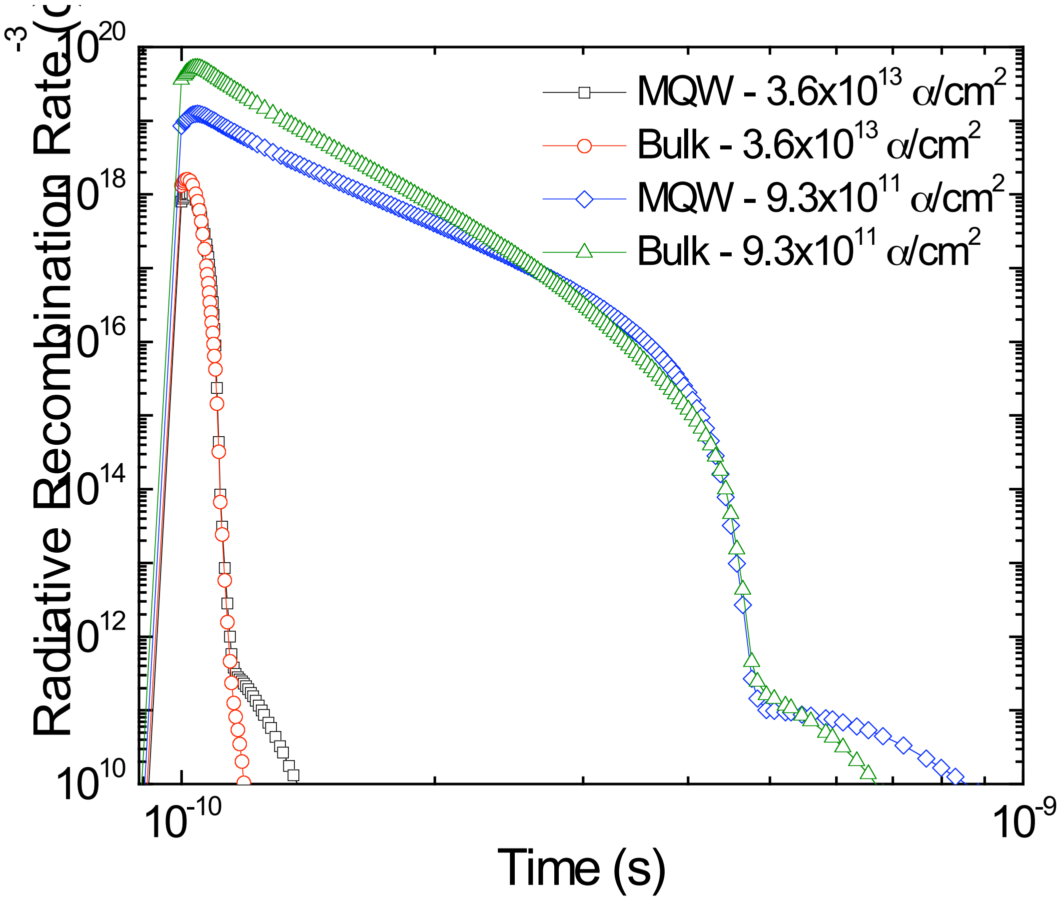


Fig. 5 Time-dependent radiative recombination rate in the MQW and bulk GaN for two fluences,$3.6\text{x}10^{13}$ alpha/cm^2 and $9.3\text{x}10^{11}$ alpha/cm^2, calculated with Silvaco Atlas. Calculations assume a capture cross section for Ga and N vacancies of $3\text{x}10^{-15}$ cm^2.

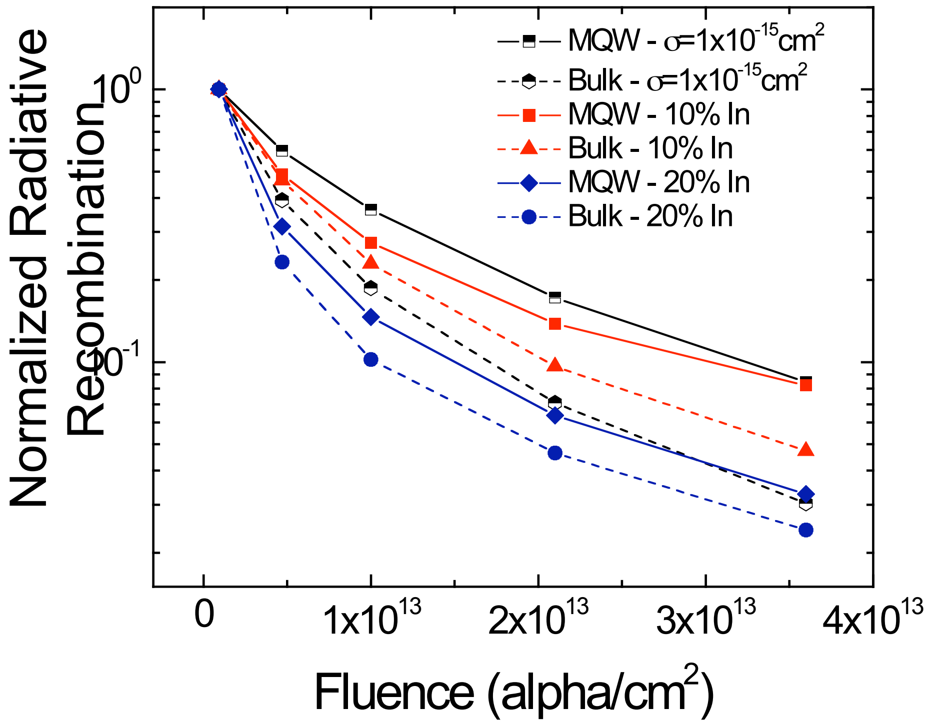


Fig. 6 Comparison of normalized measured alpha-luminescence (as shown in Fig. 1) with simulated total radiative recombination in quantum well/GaN for trap cross sections of 1×10^{-15} cm^2.